Compositing in $Yb_{14}MnSb_{11}$: a strategy to improve electronic transport

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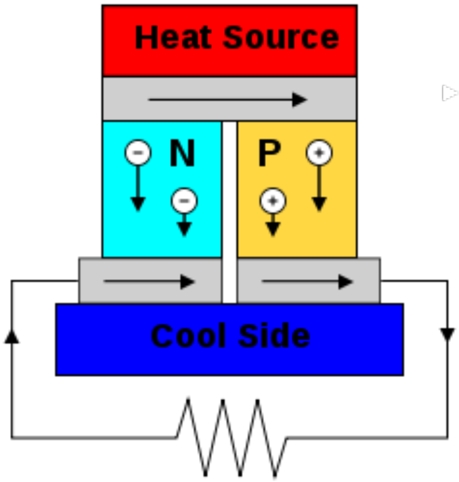
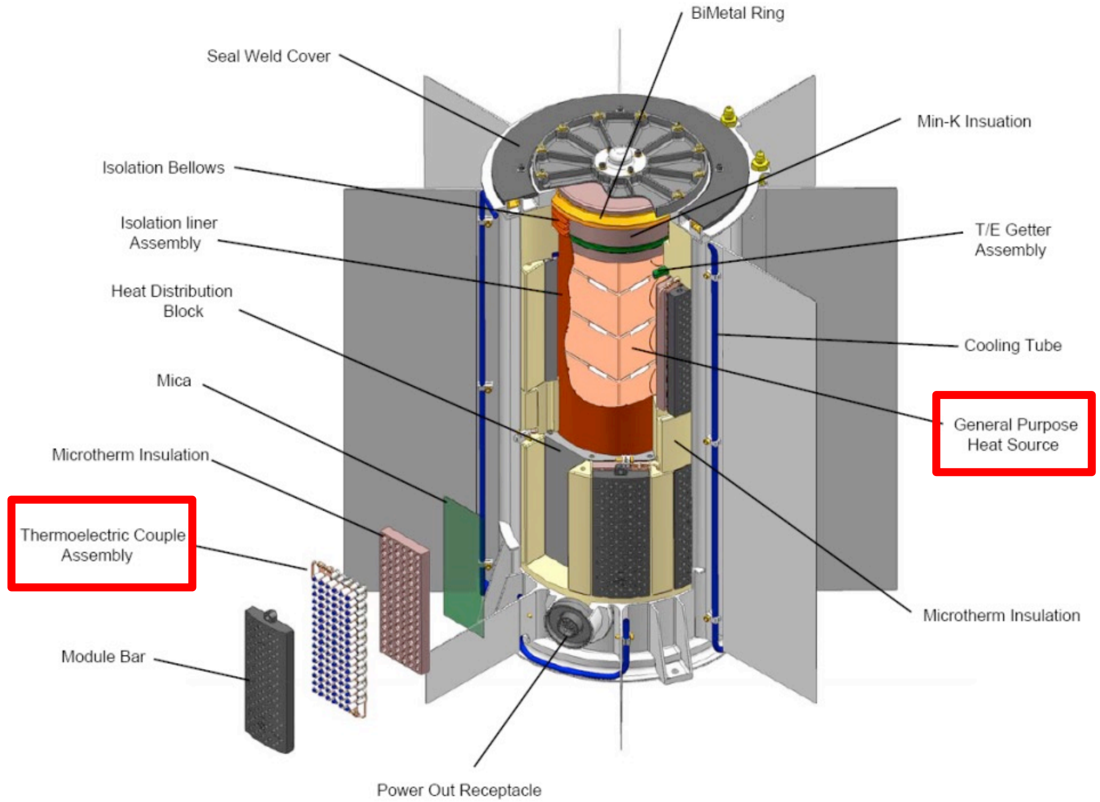
Thermoelectric generators

TE p-n junction



https://en.wikipedia.org/wiki/Thermoelectric_generator, downloaded on 9th March 2017.

https://rps.nasa.gov/resources/56/enhanced-multi-mission-radioisotope-thermoelectric-generator-emmrtg-concept/



Potential eMMRTG

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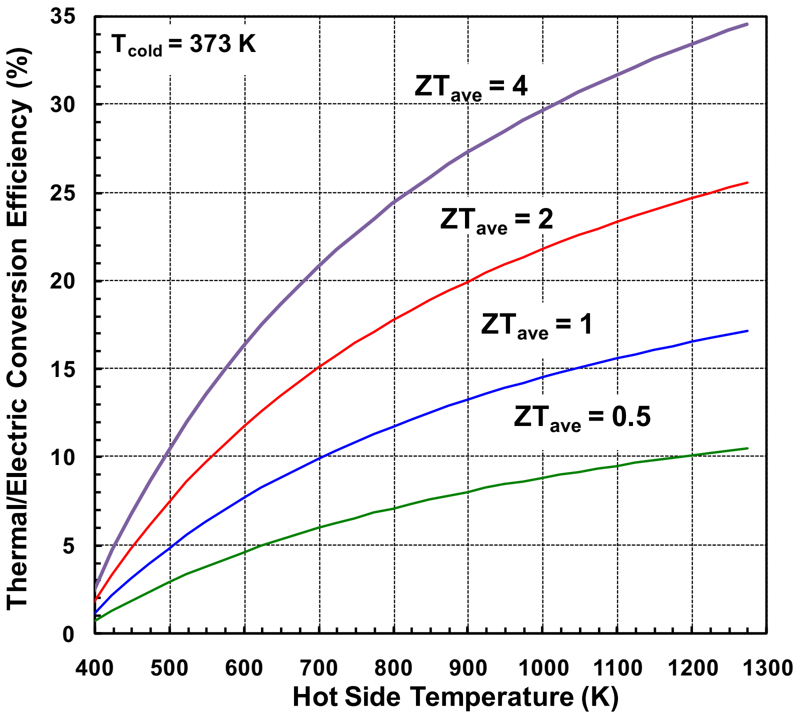
Thermoelectric power generation



Power generation

(across 1275 to 300 K)

State-Of-Practice materials:

$ZT_{average}$ ~ 0.5

State-Of-the-Art materials:

$ZT_{average}$ ~ 1.1

Best SOA materials:

ZT_{peak} ~ 1.5 to 2.0

Efficiency

Carnot | TE Materials

$$\eta_{\max} = \frac{T_{hot} - T_{cold}}{T_{hot}} \frac{\sqrt{1+ZT} - 1}{\sqrt{1+ZT} + \frac{T_{cold}}{T_{hot}}}$$

Figure of merit

$$zT = \frac{S^2 T}{\rho\kappa}$$

S = Seebeck coefficient (thermopower)

ρ = Electrical resistivity

κ = Thermal conductivity

T = Absolute temperature

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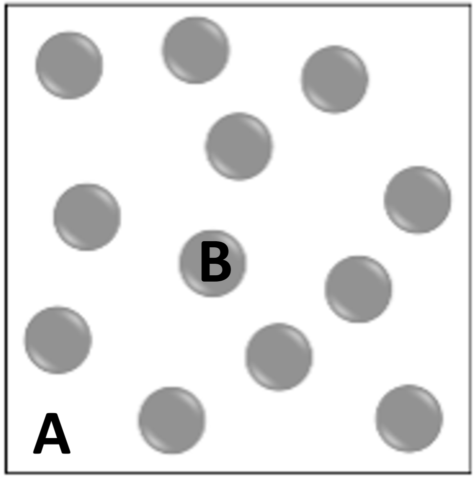
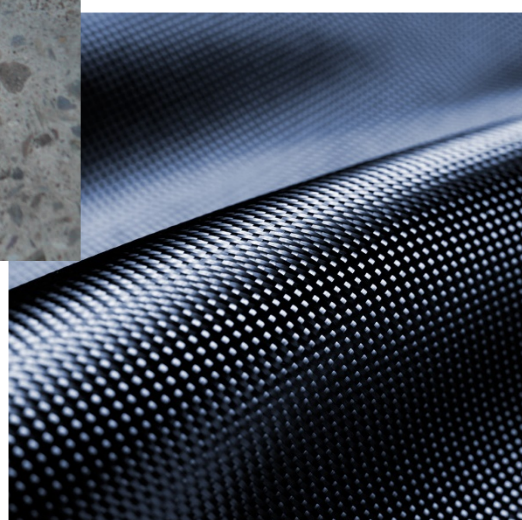
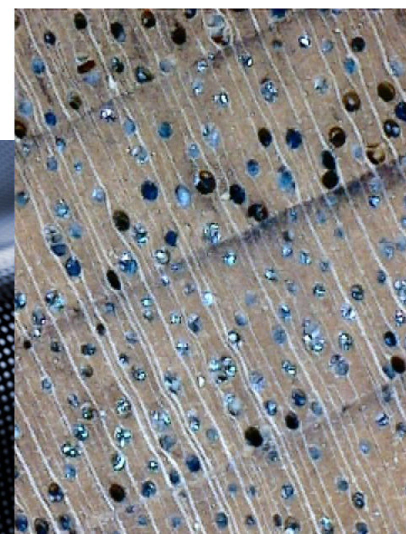
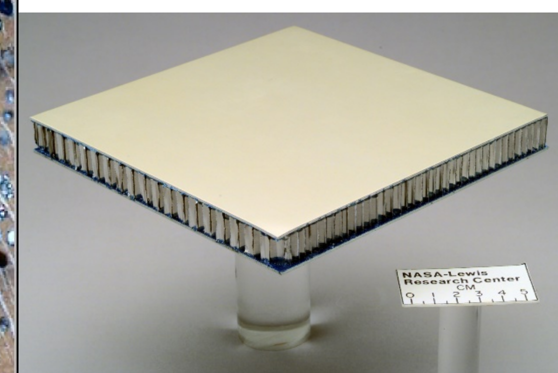
Composite



Composite material = a material made from two or more constituents with different physical or chemical properties that, when combined, produce a material with characteristics different from the individual components. The individual components remain separate and distinct within the finished structure.







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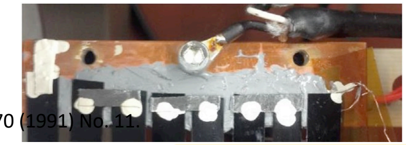
Thermoelectric composite



Bergman, Levi, J. Appt. Phys., Vol. 70 (1991) No. 11.

Organic-inorgar

coefficient α_e for a variety of situations. For the thermoelectric figure of merit Z_e of an isotropic, two-component composite, we also found exact bounds which prove that Z_e can never exceed the largest component value of Z. This

What about inorganic-inorganic composites?

Bergman, Fel, J. Appt. Phys., Vol. 85 (1999) No. 12.

We have shown that the thermoelectric power factor W_e can sometimes be enhanced by making a composite mixture of two materials. When such an enhancement is possible, the

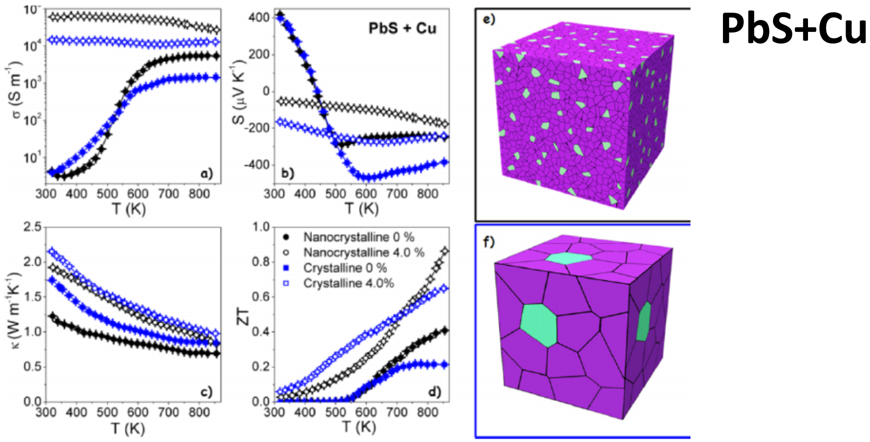
Liu et al., APL Mater. 4 (**2016**) 104813.

Thermoelectric composite

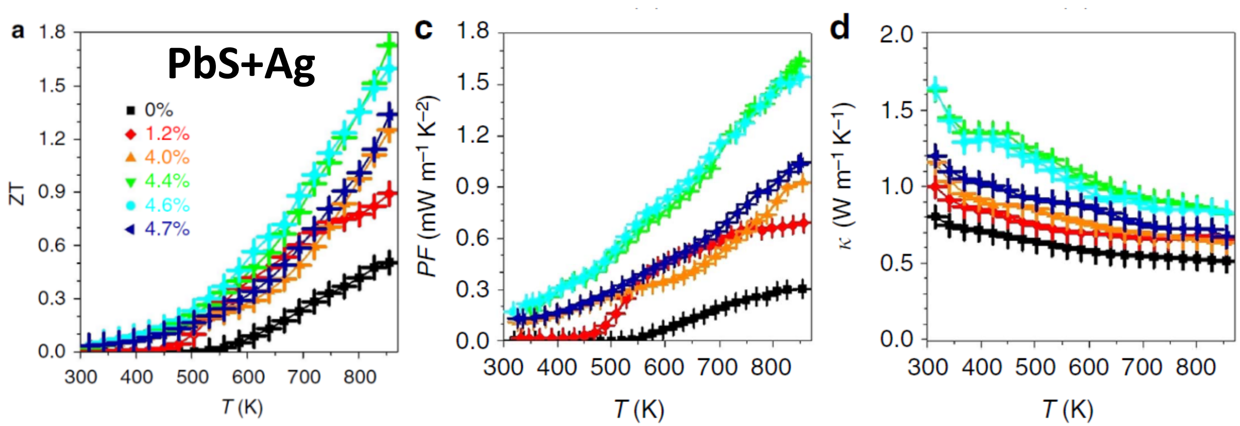
Recent studies showed it is possible to improve zT

Semiconductor	Metal	κ	S	σ	n/p	μ	Reference
PbS (N)	Ag						14
PbTe (P)	Pb						17
Bi_2Te_3 (N)	Au						18
Bi_2Te_3 (N)	Cu						19
Bi_2Te_3 (N)	Ag						20
Bi_2Te_3 (N)	Bi						21
Sb_2Te_3 (P)	Ag						22
Sb_2Te_3 (P)	Pt						22
$Bi_{0.5}Sb_{1.5}Te_3$ (N)	Cu						23
$Bi_{0.5}Sb_{1.5}Te_3$ (P)	Ag						24
$Bi_{0.5}Sb_{1.5}Te_3$ (P)	Cu						24
$Bi_2Te_{2.7}Se_{0.3}$ (N)	Pt						25
$Bi_2(Te_{0.9}Se_{0.1})_3$ (P)	Cu						26
$Bi_2(Te_{0.9}Se_{0.1})_3$ (P)	Zn						26
GaAs (N)	Bi						27
GaAs (N)	In						28
$FeSb_2$ (N)	Cu						15
$FeSb_2$ (N)	AgSb						29
$Ba_{0.3}Co_4Sb_{12}$ (N)	Ag						30
Na_xCoO_2 (P)	Au						31
$Bi_2Sr_2Co_2O_y$ (P)	Ag						32
$Ca_3Co_4O_{9+\delta}$ (P)	Ag						33
$Ca_3Co_4O_9$ (P)	Ag						34
$Na_xCo_2O_4$ (P)	Ag						35

Liu et al., APL Mater. 4 (**2016**) 104813.

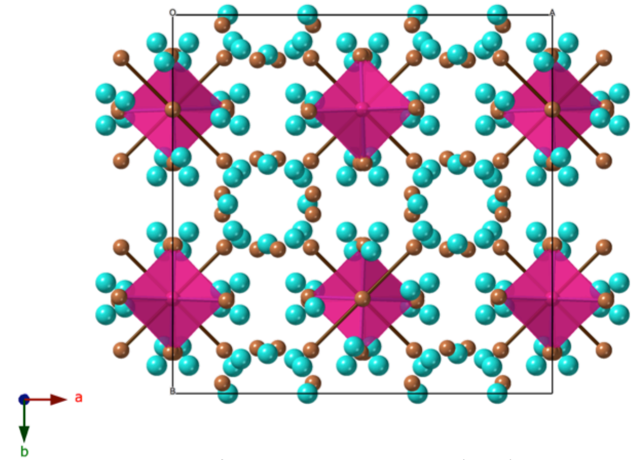


Liu et al., APL Mater. 4 (**2016**) 104813.



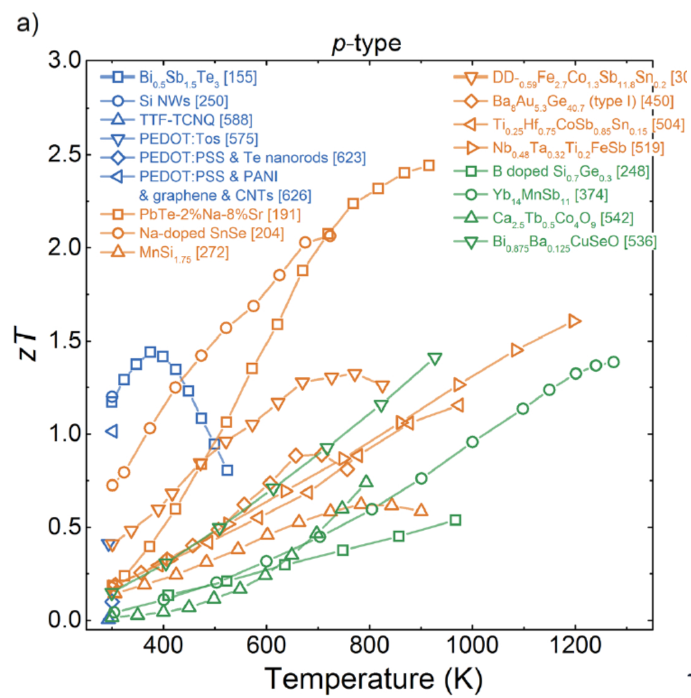
Ibañez et al., Nature Communications 7 (**2016**) 10766.

$Yb_{14}MnSb_{11}$



Journal of Solid State Chemistry, 271 (**2019**) 88–102

- ***$Yb_{14}MnSb_{11}$***
  - Zintl Structures
  - Covalent, anionic substructures: $[MPn_4]^{9-}$, $[Pn_3]^{7-}$, $4Pn^{3-}$, $14A^{2+}$
  - Body centered tetragonal ($I4_1/acd$)
  - 208 atoms per unit cell
  - P-type conductor
- ***Low thermal conductivity*** (~0.85 $Wm^{-1}K^{-1}$)
- ***Peak zT ~1.3 @1273K***
- ***3x improvement over SiGe***



Material Science and Engineering R (**2018**). DOI:10.1016/j.mser.2018.09.001

Composite metal (M) choice

$Yb_{14}MnSb_{11}$ + M

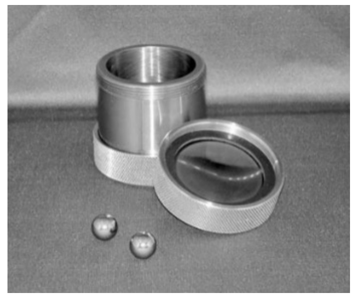
Inclusions	M-Sb Reactivity	ρ [nΩ·m] @25°C	E [GPa]	CTE [um/mK] @25°C
Ni	High	69.3	200	13.4
Co	Low	62.4	209	13
W	None	52.8	411	4.5

- Ni reacted with matrix forming secondary phases → not good.
- We used Co →  **Co**balt **Co**mposite **N**etwork **U**sing **T**hermoelectrics (**CoCoNUT**)
- We used W → Journal of Applied Physics (DOI: 10.1063/1.5118227)

$Yb_{14}MnSb_{11}$ + M synthesis

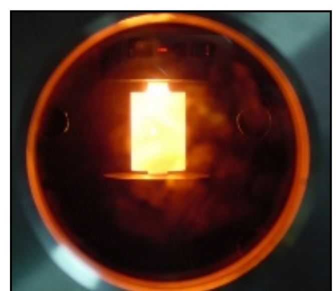
$Yb_{14}MnSb_{11}$ | +2vol%Co | +5vol%Co | +10vol%Co

Mix precursors (Yb, MnSb, Sb + CoSb/W)

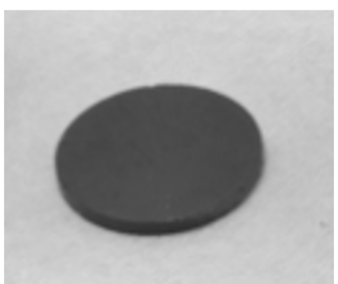


High energy ball mill

Homogenized powder

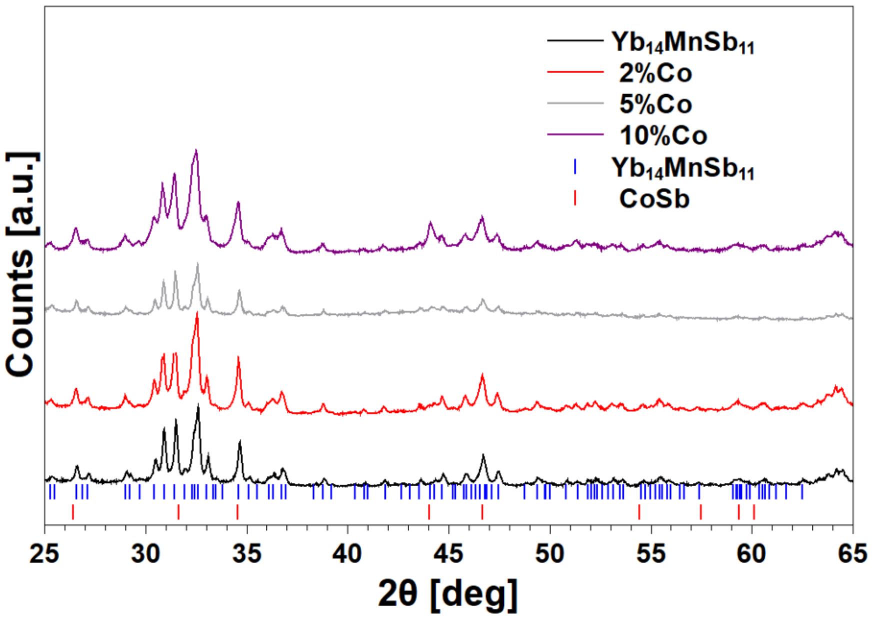


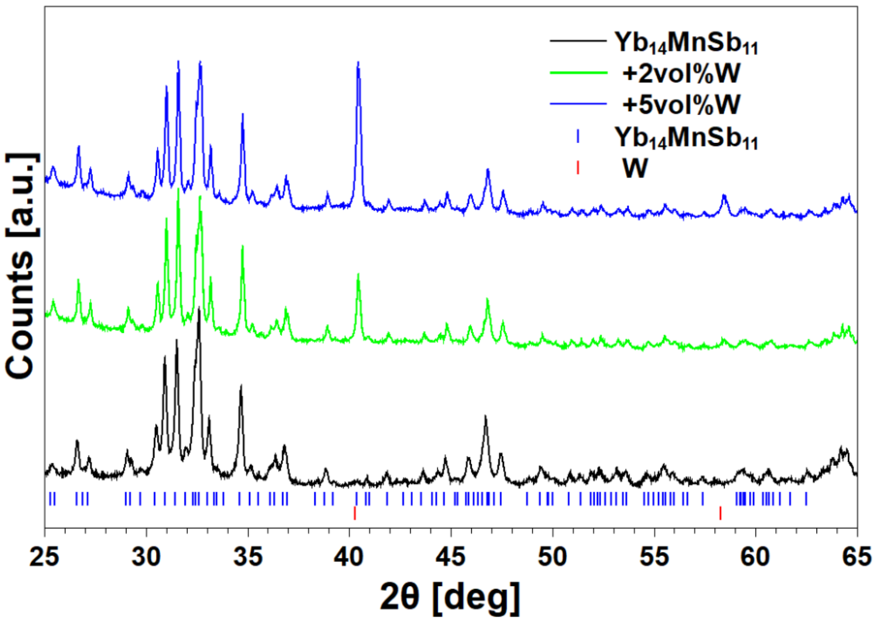
SPS Synthesis/compaction



½" pellets >98% density

XRD analysis





- Profile of each sample matches with $Yb_{14}MnSb_{11}$.
- Sample with 10%Co shows CoSb Impurities.
- W reflections are visible in 2vo%, 5vol% W samples.
- W composite samples show no impurities.

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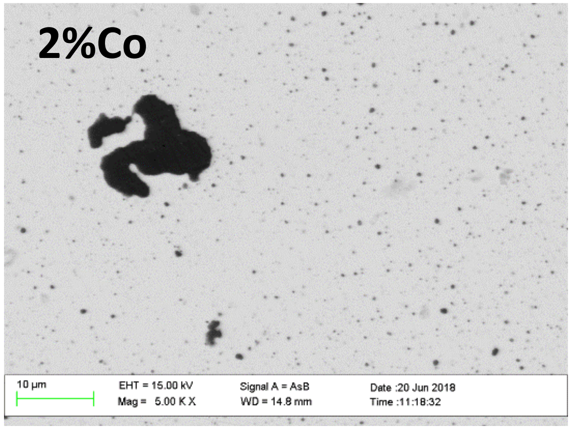
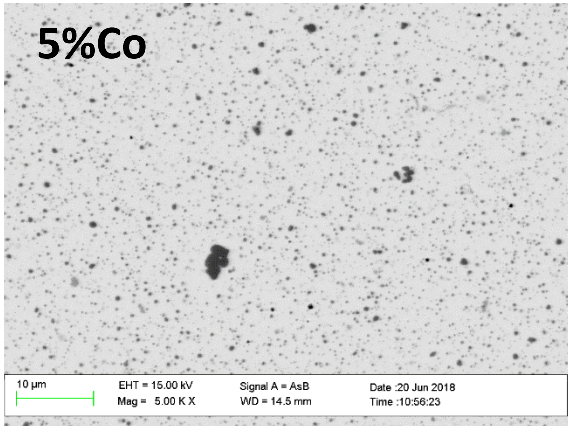
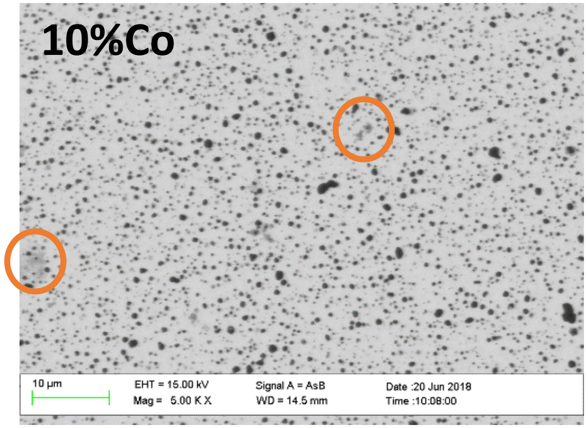
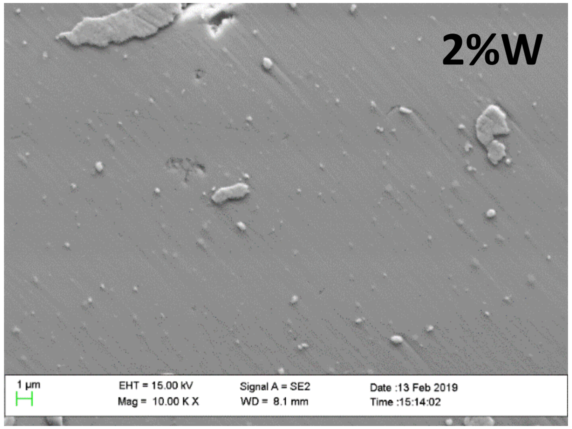
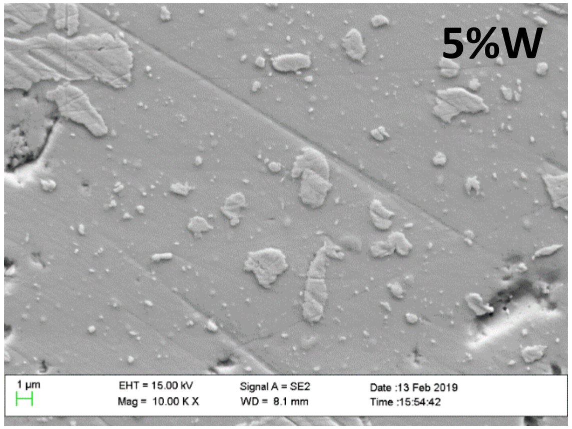
Microstructure





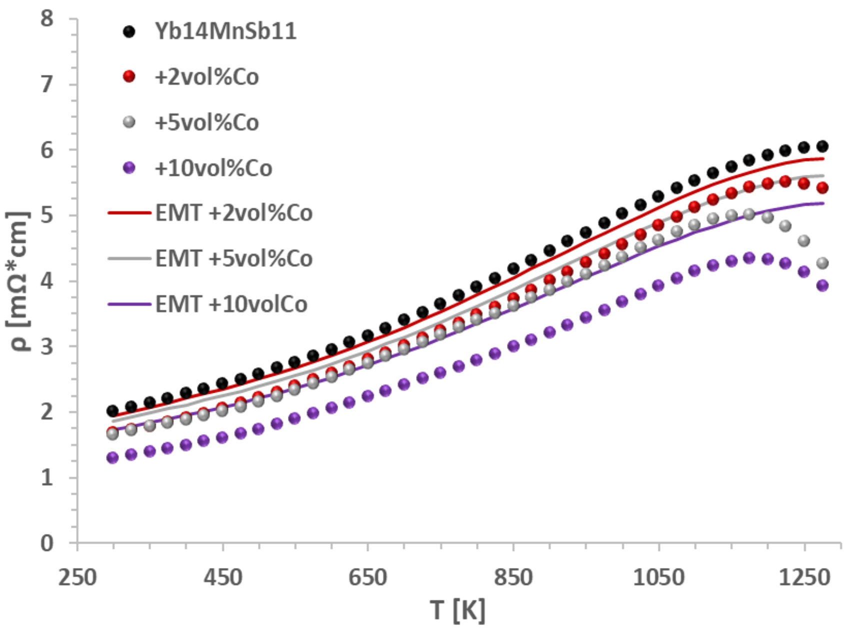


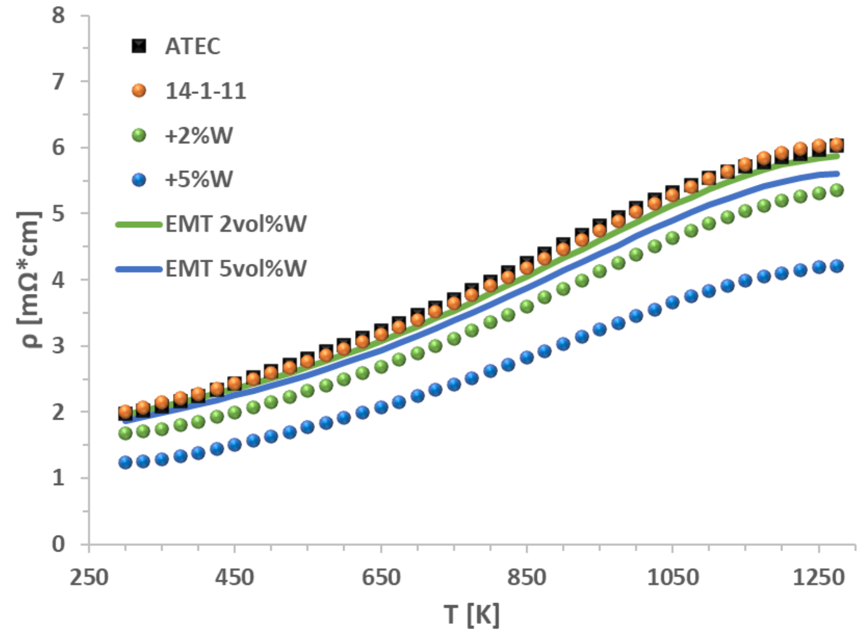




- Inclusions sizes between nm and several µm (for both Co & W).
- Signs of CoSb in 10%Co sample (not dissolved CoSb).
- No cracks radiating from inclusions (CTE mismatch).

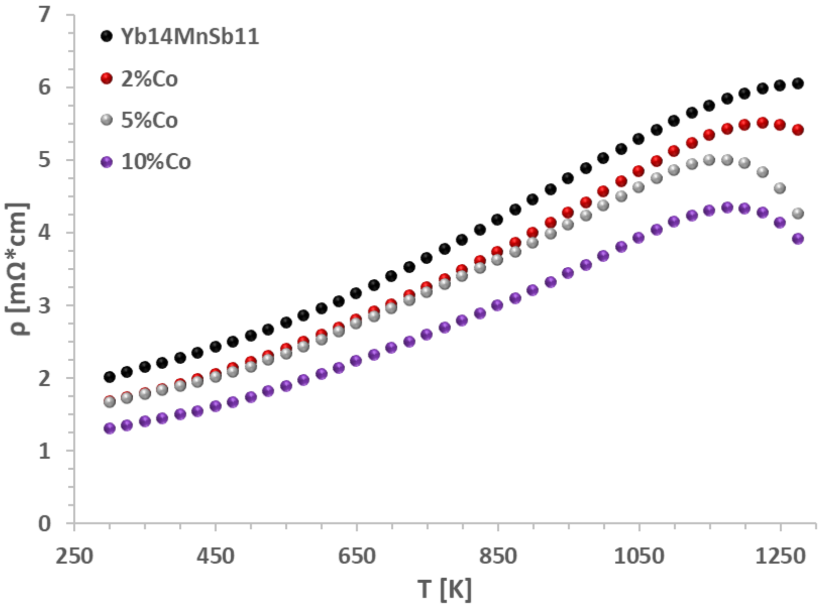
Electronic transport

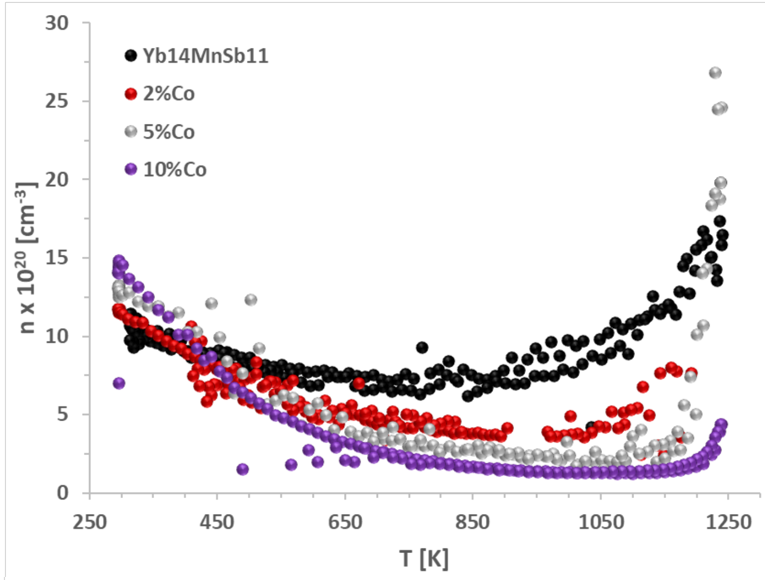


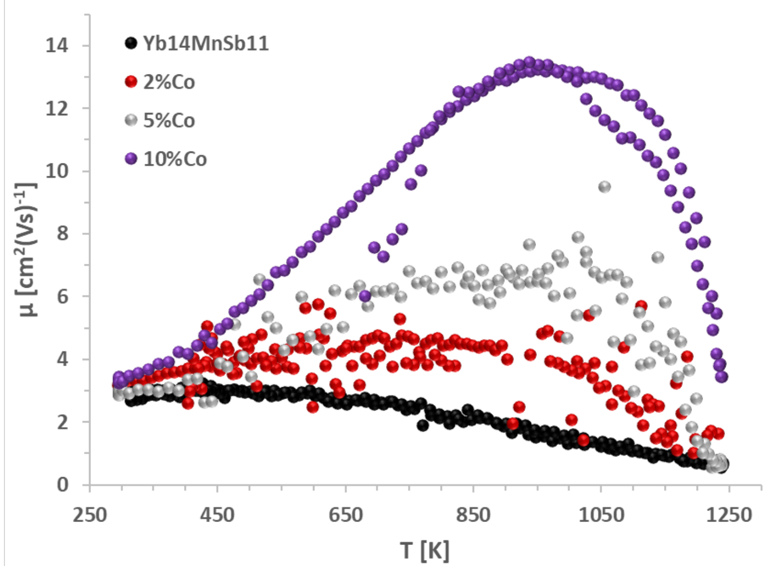


- More M → more conductive → ***ρ*** decreases (as expected).
- Similar trend for both TM.
- Effects of inclusions is more evident in 14-1-11 than in LaTe (15vol% is needed).

Electronic transport

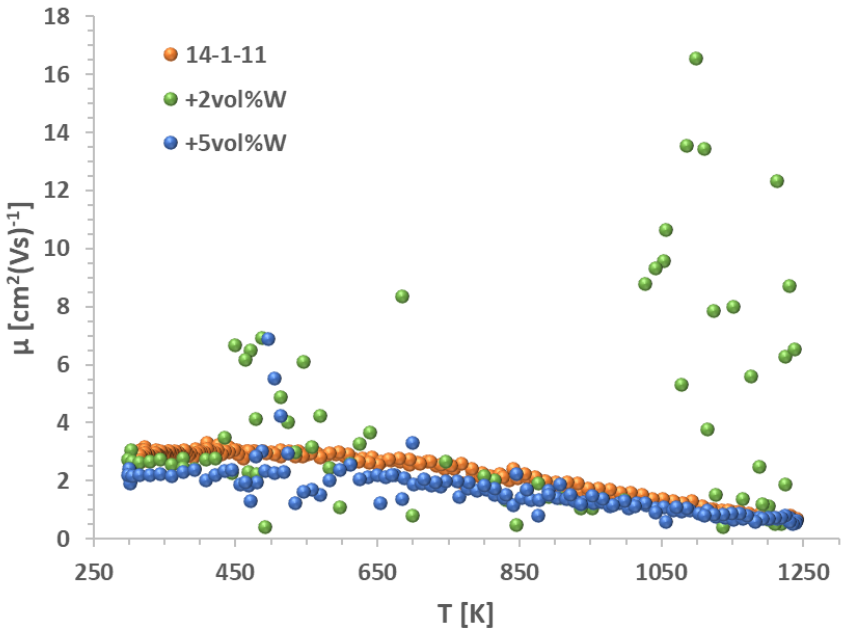


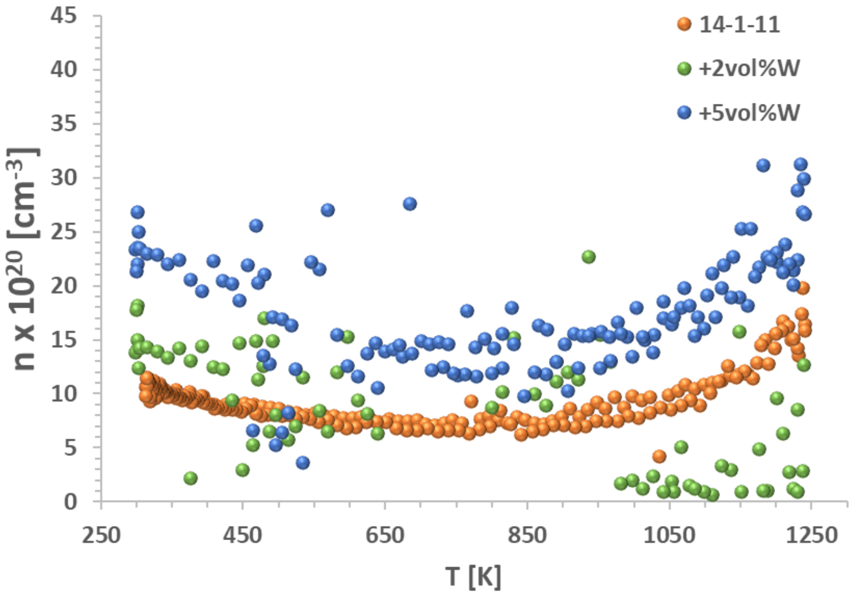




- More Co → more conductive → $\boldsymbol{\rho}$ decreases (as expected).
- Clear ferromagnetic behavior of $\boldsymbol{\mu}$ and $\boldsymbol{n}$.
- Ferromagnetic-paramagnetic transition behavior at 1200-1250 K.
- Interesting fact: F-P transition 150-200 K before Curie temperature (~1400K).

Electronic properties

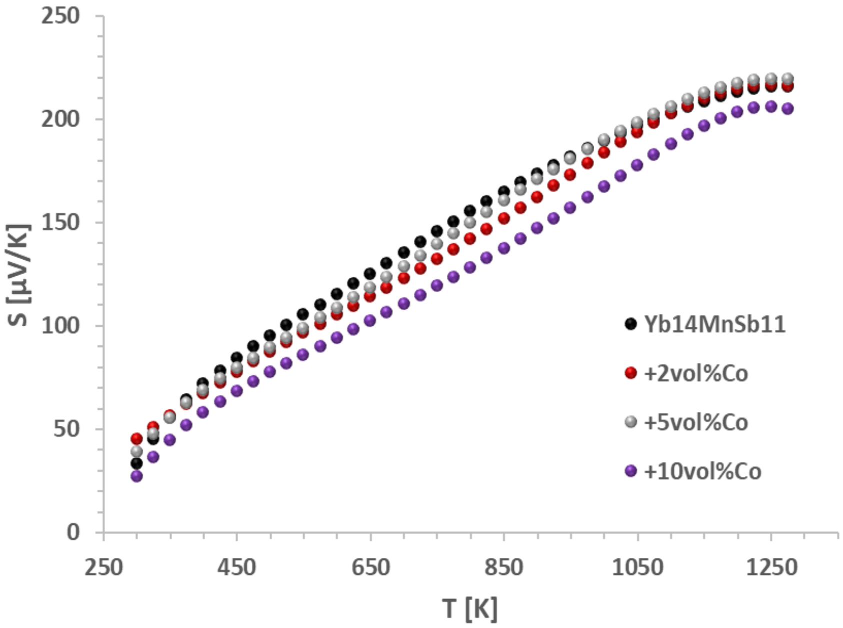


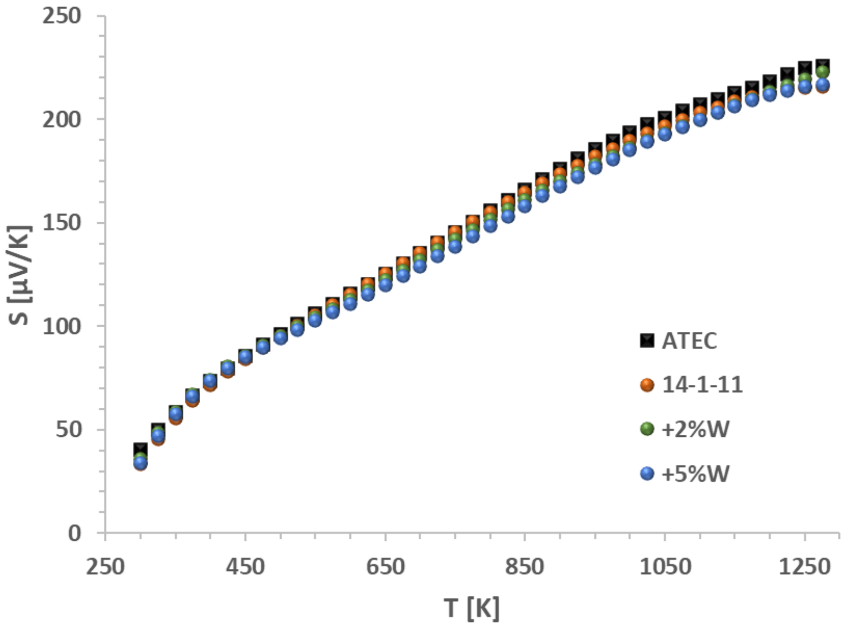


- Mobility does not change much, comparable with pristine 14-1-11.

- There's an increase of n, although data for 2vol% are noisy.

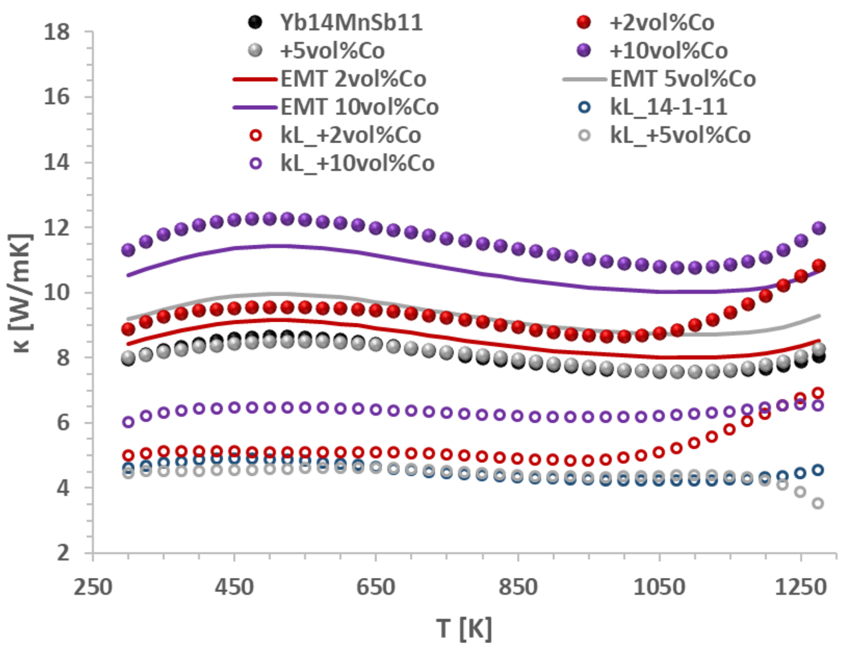
Electronic transport

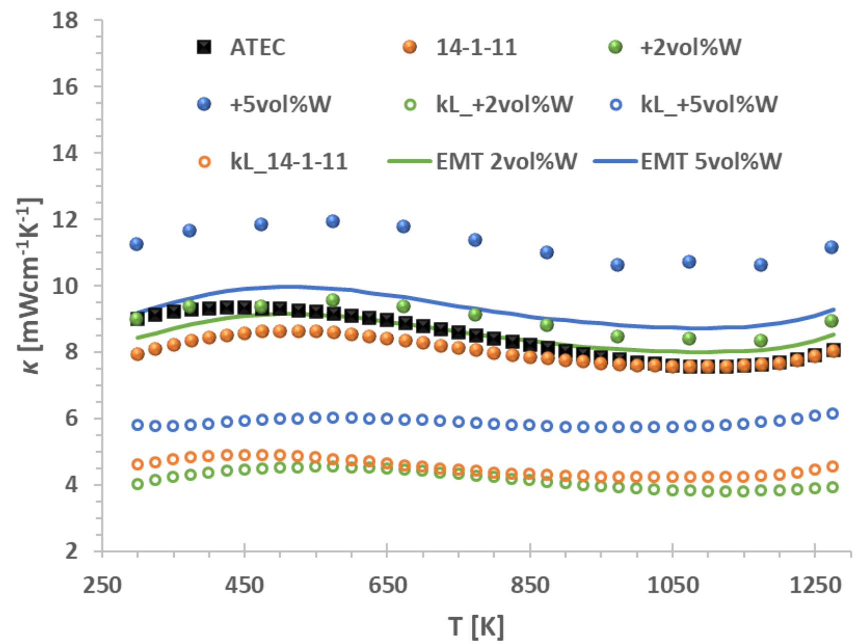




- Lower electrical resistivity should be paired with lower Seebeck coefficient.
- However, ***S*** remains similar to pristine $Yb_{14}MnSb_{11}$.
- Only sample with 10%Co experiences decrease in Seebeck coefficient.

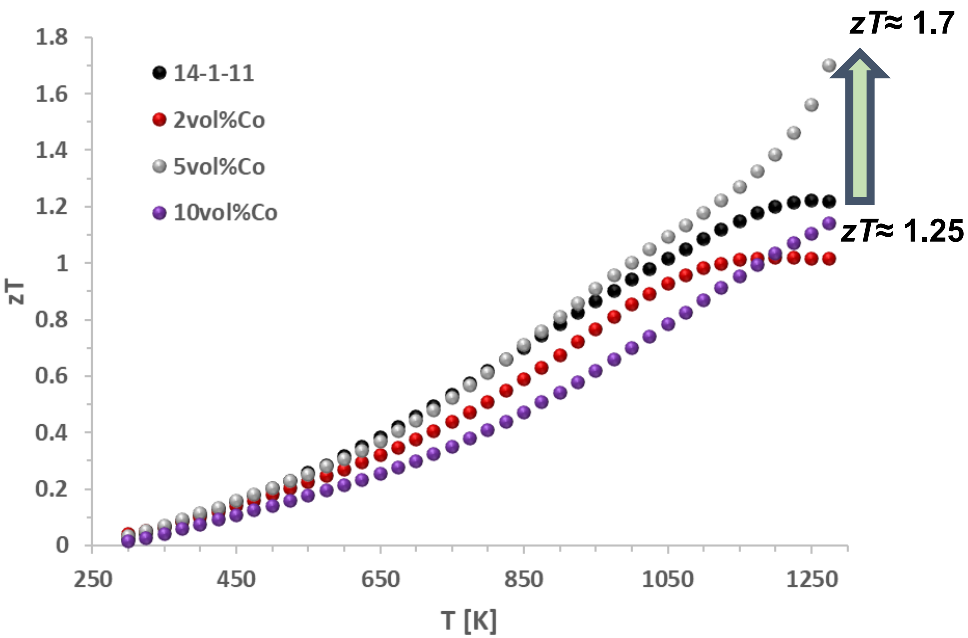
Thermal properties

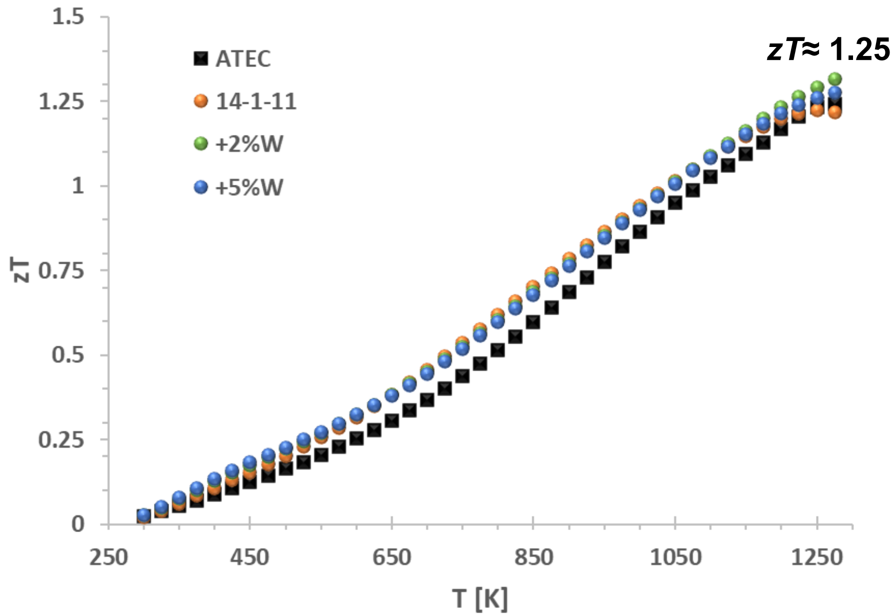




- C_p corrected to include inclusion contribution.
- Up to 5% Co inclusions, thermal conductivity is comparable to pristine $Yb_{14}MnSb_{11}$.
- In W samples 5% already have higher $\boldsymbol{\kappa}$.

Thermoelectric performance





- Increase PF
- κ comparable up to 5% Co.
- In the end zT improves from 1.25 to 1.7.

- Increase PF canceled out from increase κ.
- In the end zT does not vary.

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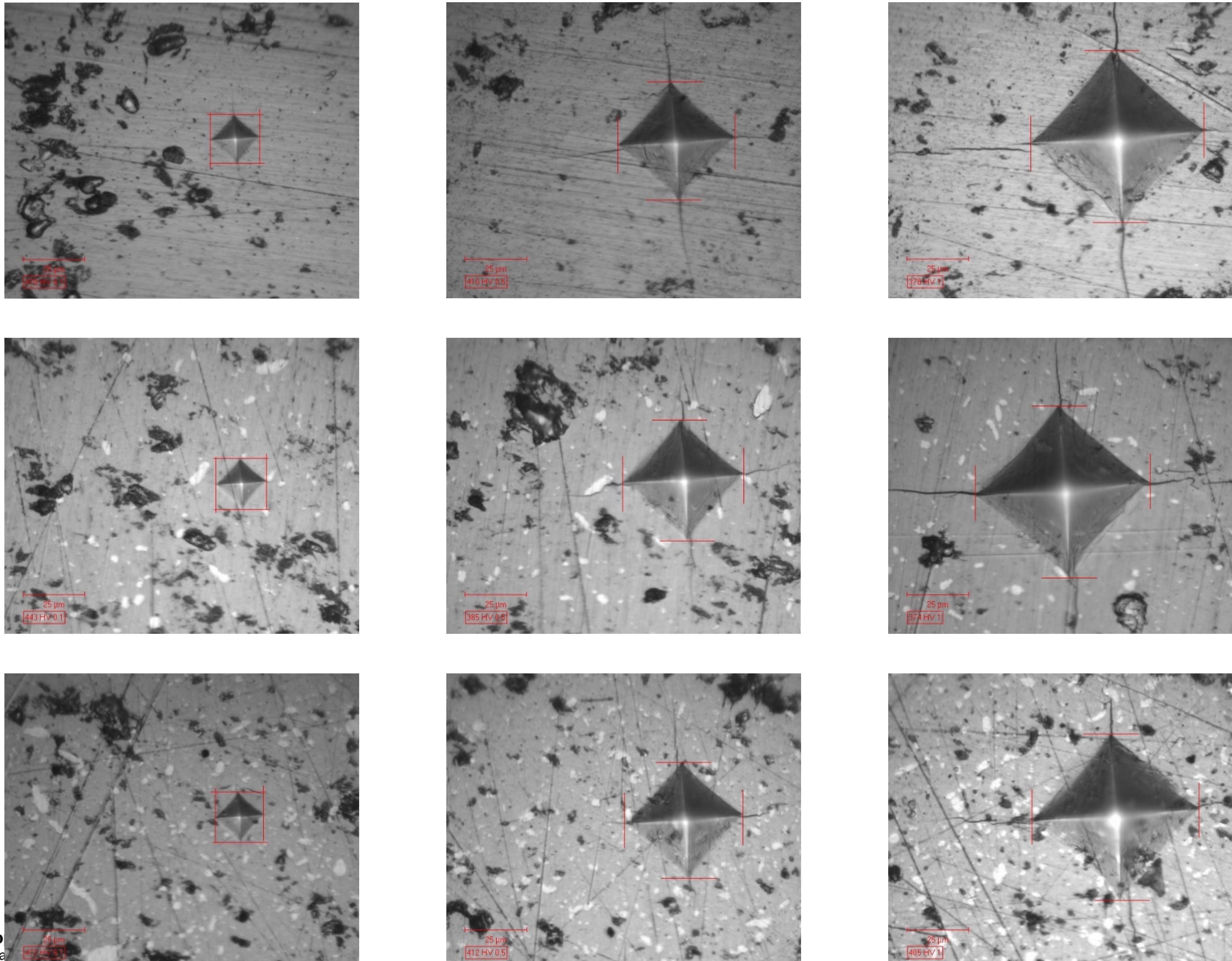
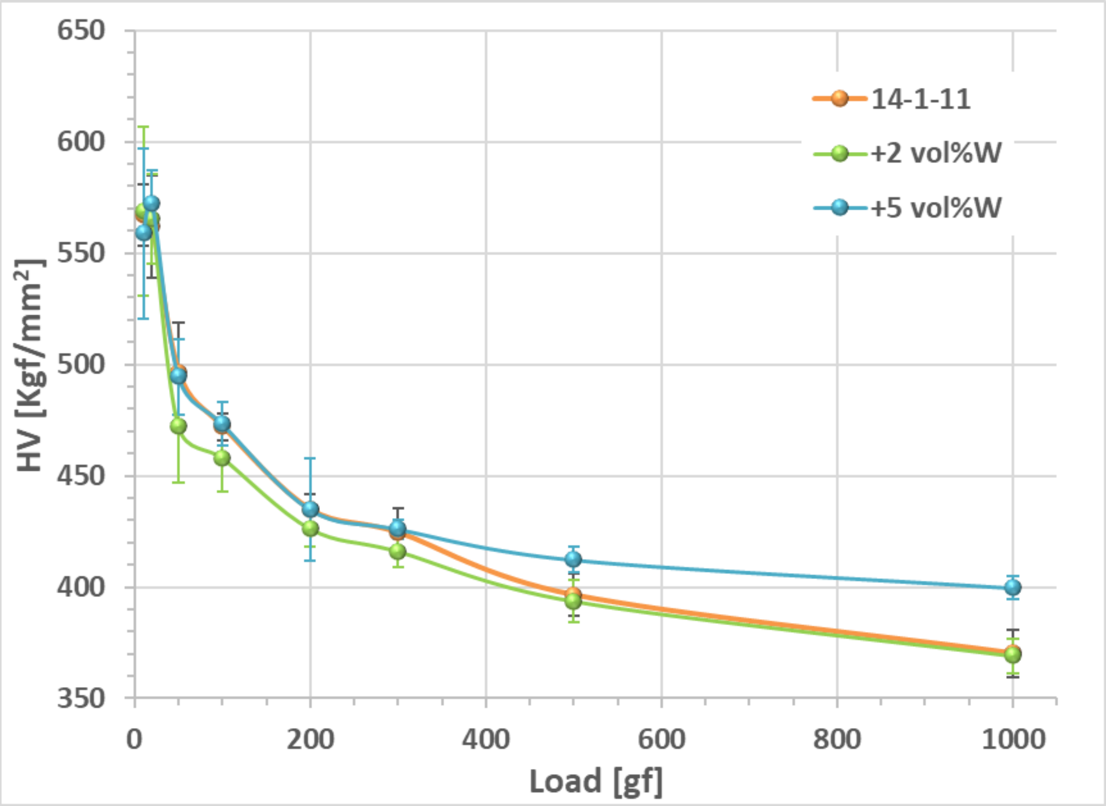
	100g	500g	1Kg
14-1-11			
2vol% W			
5vol% W			



Vickers Hardness



- At low loads (up to 300gf) the HV values are comparable for all samples.

- The last two loads (0.5 and 1 Kgf) the sample with 5 vol%W shows higher HV.

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Acknowledgments

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- **TECT Group, JPL**
- **Power and Sensors Systems Section, JPL**

QUESTIONS??

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